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Effects of enhancing nitrogen use efficiency in cropland and livestock systems on agricultural ammonia emissions and particulate matter air quality in China

Biao Luo et al.

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Figure S1–S5
Table S1–S12

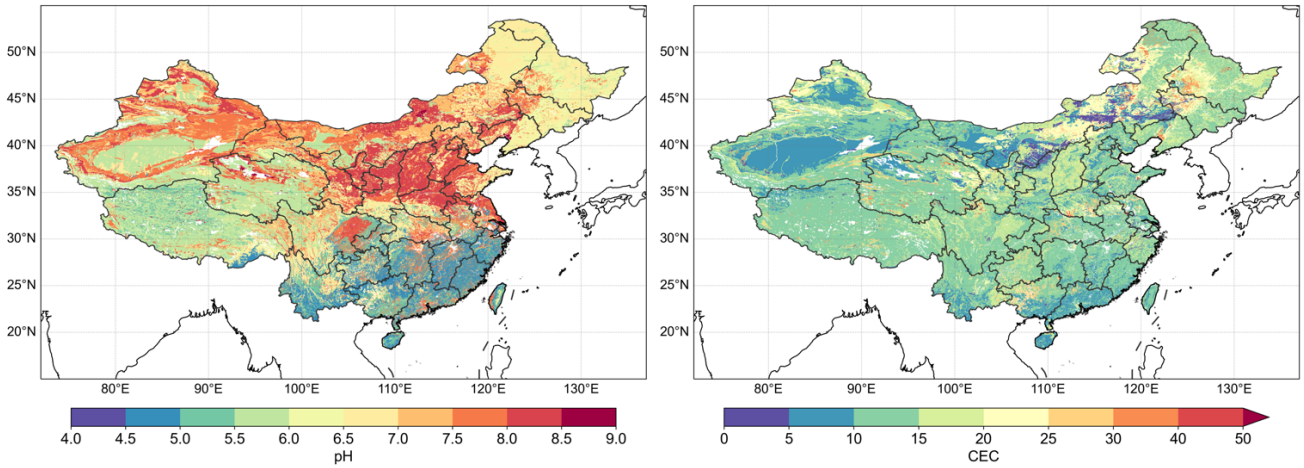

Figure S1. Soil pH and cation exchange capacity (CEC) of China

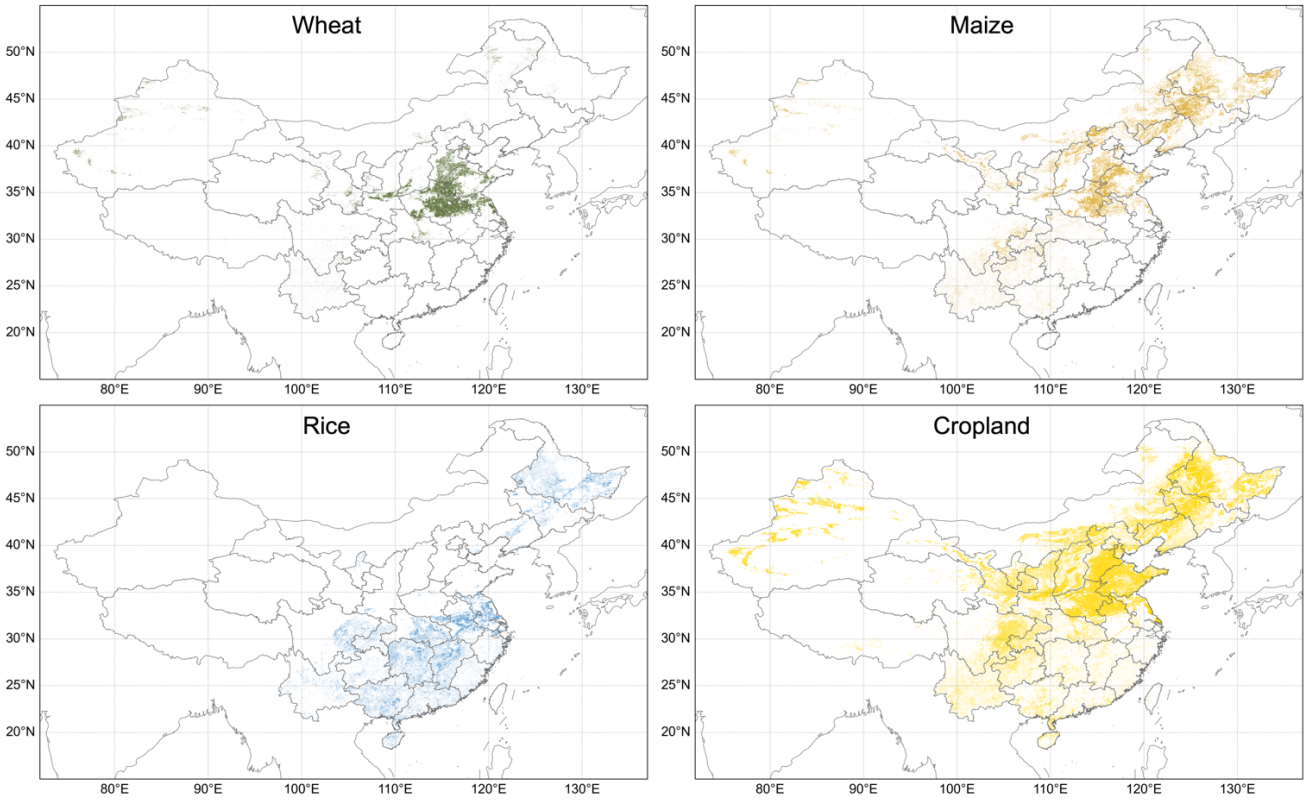

Figure S2. Spatial distribution of wheat, maize, rice and croplands

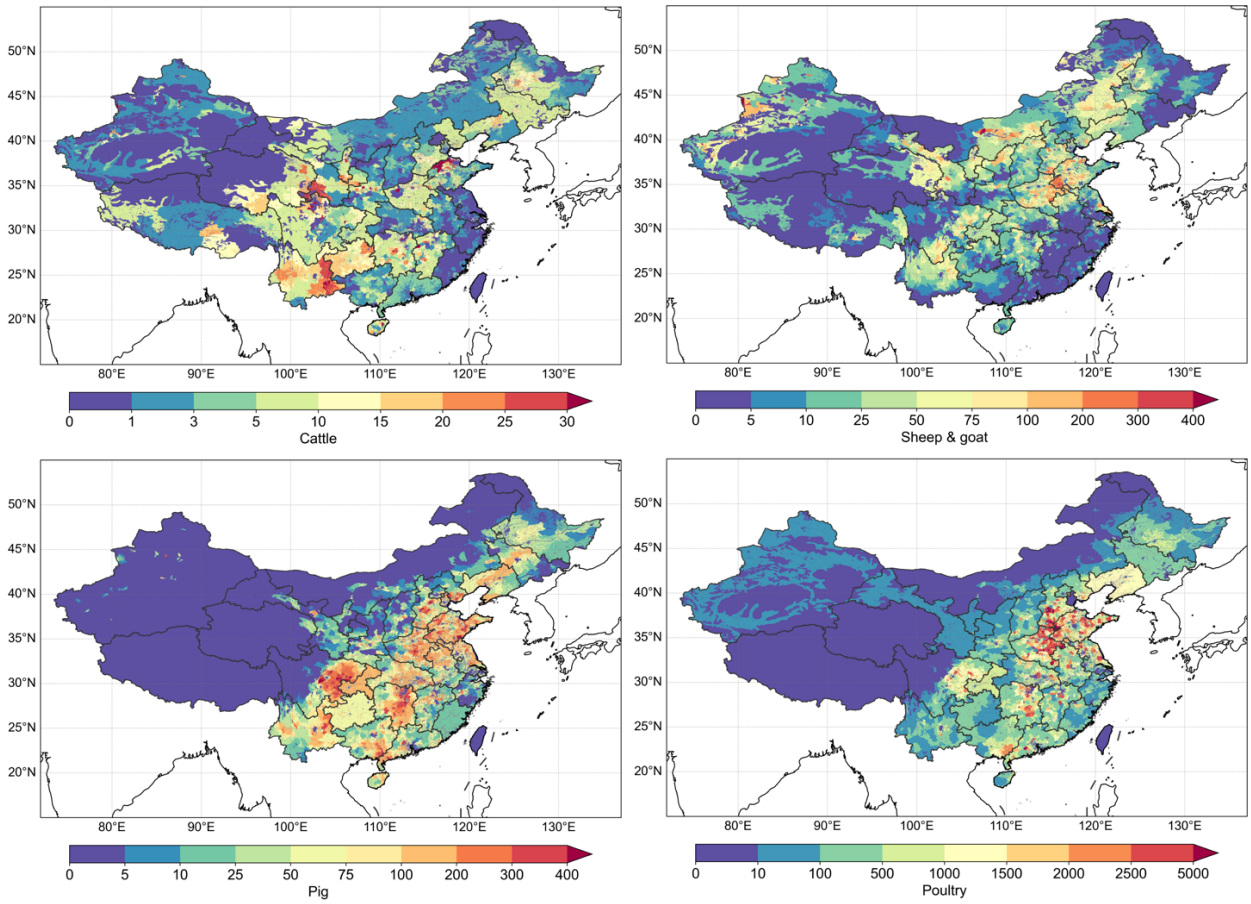

Figure S3. Spatial distribution of cattle, sheep & goat, pig and poultry (head per grid)

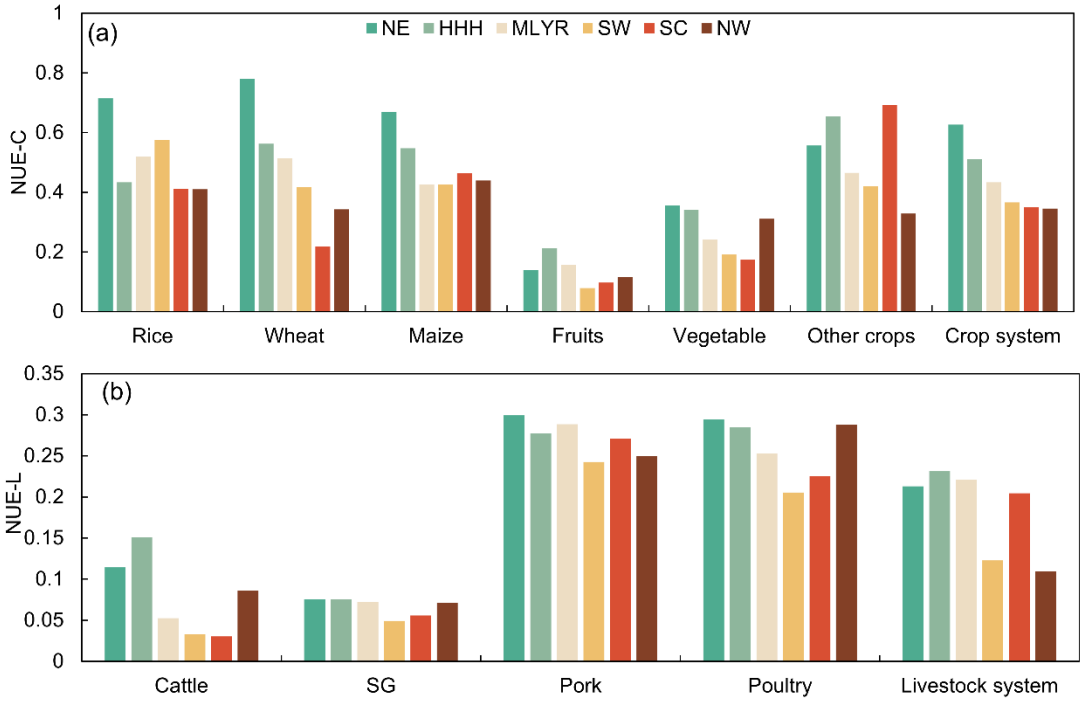

Figure S4. NUE-C **(a)** and NUE-L **(b)** of different agricultural sub-regions

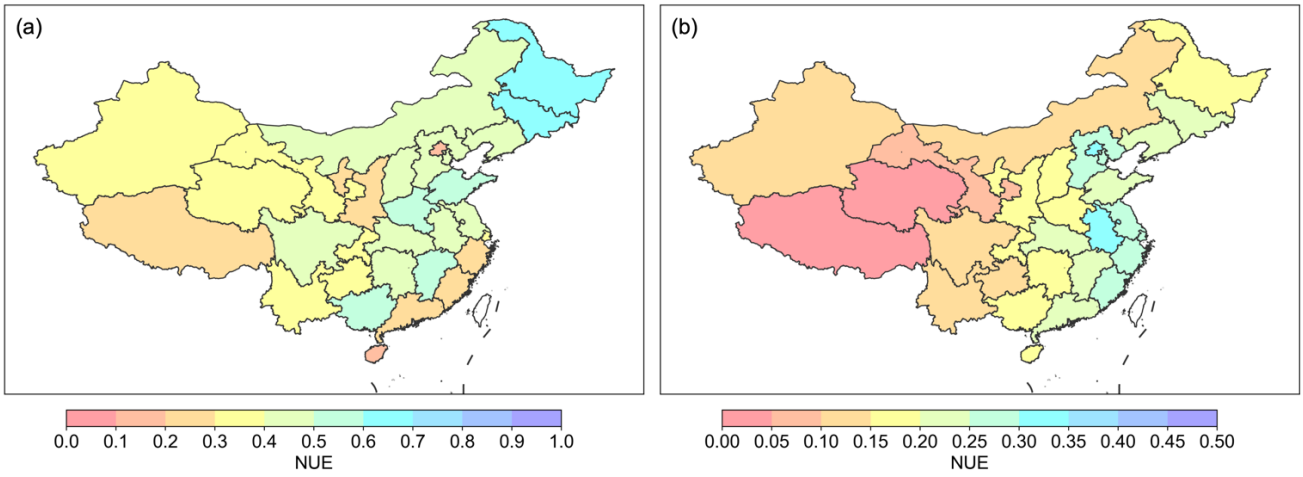

Figure S5. NUE for crop system (**a**) and NUE for livestock system (**b**)

Table S1.N fertilizer application dates and rates for main crops in China

Crops	N fertilizer application rate (kg N ha^{-1})[a]	Application day (calendar day)[b]					
		First	Second	Third	Fourth	Fifth	Deviation
Spring wheat	222.70	90	110	135			29
Winter wheat	222.70	270	300	73			27
Maize	198.45	120	135	145	160		36
Early rice	173.3	100	110	120	135	170	32
Middle rice	170.45	120	130	140	155	190	32
Late rice	170.65	180	190	200	215	250	32
Potato	144.00	132	149	157			23
Sweet potato	104.00	115	125	175	195		17
Soybean	49.65	120	136	191			32
Oilseed rape	138.60	275	305	84	91		30
Sunflower	131.15	95	105	125			20
Peanut	123.20	130	150				22
Cotton	294.35	105	119	159	205	215	24
Beets	171.00	110	180	250			20
Sugar cane	455.95	60	90	170	230		20
Vegetables	276.40						
Fruits	311.50						
Tobacco	135.20	90	99	109	132		9

[a] application rates derived from National Agricultural Cost-Benefit Information Compendium (NBSC, 2023); [b] derived from Zhang et al., (2018) and agricultural resources atlas, http://geo.ckcest.cn/dlsagri/, last access: December 2022.

Table S2. Emission factor for fertilizer application (derived from Zhang et al. (2018))

Factor	Value
Crop type	
Upland crops	–0.045
Rice	0
Fertilizer type	
Ammonium sulfate	0.429
Urea	0.666
Ammonium nitrate	–0.35
Compound NPK	0.014
Application mode	
Broadcast	–1.305
Injection	–1.895
Soil pH	$0.067\times pH^2–0.69\times pH+0.68$
Soil CEC	
CEC ≤ 16	0.088
16＜CEC≤24	0.012
24＜CEC≤32	0.163
CEC＞32	0

Table S3. Livestock excretion rates (derived from Huang et al. (2012))

	Excrement (kg day^{-1} cap^{-1})		Nitrogen contents (%)		Total ammoniacal nitrogen (TAN) content (%)
	Urine	Feces	Urine	Feces	
Cattle	5	7	0.9	0.38	60
Horse	6.5	15	1.4	0.2	60
Goat	0.66	1.5	1.35	0.75	60
Sheep	0.66	1.5	1.35	0.75	60
Pork	3.2	1.3	0.4	0.34	70

Poultry	0	0.1	0	1.1	70

Table S4. Parameters for estimating ammonia emissions from livestock waste management (derived from Huang et al. (2012) and Kang et al. (2016), %)

Parameters	X_slurry	EF_outdoor	EF_house_slurry			EF_house_solid			EF_storage_slurry				EF_storage_solid				EF_spread_slurry	EF_spread_solid
			T<10℃	10–20℃	T>20℃	T<10℃	10–20℃	T>20℃	NH_3	N_2O	NO	N_2	NH_3	N_2O	NO	N_2		
Free-range																		
Cattle	11	53	4.7	7	9.3	4.7	7	9.3	20	1	0.01	0.3	27	8	1	30	55	79
Horse	11	0	9.3	14	18.7	9.3	14	18.7	35	1	0.01	0.3	35	8	1	30	90	81
Goat	11	53	4.7	7	9.3	4.7	7	9.3	20	1	0.01	0.3	28	8	1	30	55	80
Sheep	11	53	4.7	7	9.3	4.7	7	9.3	20	1	0.01	0.3	28	8	1	30	55	80
Pork	11	0	9.5	15.6	21.7	9.5	15.6	21.7	14	1	0.01	0.3	45	5	1	30	40	81
Poultry	11	58.5	23.1	42.3	52.4	23.1	42.3	52.4	0	0	0	0	20	3	1	30	0	63
Intensive																		
Cattle	50	53	4.7	7	9.3	4.7	7	9.3	15.8	1	0.01	0.3	4.2	8	1	30	55	79
Horse	50	0	9.3	14	18.7	9.3	14	18.7	15.8	1	0.01	0.3	4.2	8	1	30	90	81
Goat	50	53	4.7	7	9.3	4.7	7	9.3	15.8	1	0.01	0.3	4.2	8	1	30	55	80
Sheep	50	53	4.7	7	9.3	4.7	7	9.3	15.8	1	0.01	0.3	4.2	8	1	30	55	80
Pork	50	0	9.5	15.6	21.7	9.5	14	21.7	3.8	1	0.01	0.3	4.6	5	1	30	40	81

Poultry	50	58.5	0	0	0	20.9	38	38	0	0	0	0	2.2	3	1	30	0	63
Grazing																		
Cattle	0	6	4.7	7	9.3	4.7	7	9.3	20	1	0.01	0.3	27	8	1	30	55	79
Horse	0	35	9.3	14	18.7	9.3	14	18.7	35	1	0.01	0.3	35	8	1	30	90	90
Goat	0	6	4.7	7	9.3	4.7	7	9.3	24	1	0.01	0.3	27	8	1	30	55	79
Sheep	0	6	4.7	7	9.3	4.7	7	9.3	24	1	0.01	0.3	27	8	1	30	55	79
Pork	0	0	0	0	0	0	0	0	0	1	0.01	0.3	0	5	1	30	0	0
Poultry	0	0	0	0	0	0	0	0	0	0	0	0	0	3	1	30	0	0

Note: Both solid and slurry forms exist in the management of livestock excreta. X_slurry represents the ratio of slurry in total excreta. EF_outdoor means the emission factor of ammonia loss outside the house (% of total TAN). EF_house_slurry and EF_house_solid are the emission factors of ammonia loss in the housing stage for manure handled as slurry and solid (expressed as the percentage of TAN, %). EF_storage_slurry and EF_storage_solid are the emission factors of ammonia loss in the storage stage for manure handled as slurry and solid (expressed as the percentage of TAN, %). EF_spread_slurry and EF_ spread_solid are the emission factors of ammonia loss in the application stage for manure handled as slurry and solid (expressed as the percentage of TAN, %).

Table S5. Crop planting area (1000 ha) and total nitrogen fertilizer use (10000 ton) for different provinces of China in 2017

	Spring wheat	Winter wheat	Spring maize	Summer maize	Early rice	Middle rice	Late rice	Potato	Sweet potato	Soybean	Rapeseed	Sunflower	Peanut	Cotton	Beet	Sugarcane	Vegetables	Fruits	Tabaco	Others	Total nitrogen fertilizer use
Beijing	0.1	11.2	0.0	49.7	0.0	0.1	0.0	0.0	1.1	2.2	0.0	0.7	1.4	0.0	0.0	0.0	40.3	47.7	0.0	14.1	5.1
Tianjin	13.0	95.7	0.0	201.4	0.0	30.5	0.0	1.7	1.2	3.4	0.2	3.6	1.5	20.7	0.0	0.0	49.3	31.5	0.0	17.3	9.1
Hebei	9.3	2364.1	0.0	3544.1	0.0	75.0	0.0	162.8	48.8	70.1	24.5	62.1	266.8	220.6	12.2	0.0	748.6	560.0	1.3	771.3	177.4
Shanxi	0.0	560.5	0.0	1806.9	0.0	0.8	0.0	168.1	16.6	130.8	18.8	31.8	5.8	2.9	0.1	0.0	169.9	359.5	1.5	663.1	48.5
Inner Mongolia	673.9	0.0	3716.3	0.0	0.0	122.2	0.0	432.1	0.1	989.0	309.6	713.1	23.3	0.0	82.7	0.0	218.7	59.8	1.3	1731.9	120.6
Liaoning	3.6	0.0	2692.0	0.0	0.0	492.7	0.0	63.5	26.9	74.3	0.8	4.1	271.7	0.0	2.0	0.0	308.6	350.7	7.4	224.7	79.2
Jilin	2.4	0.0	4164.0	0.0	0.0	820.8	0.0	60.1	2.0	220.2	0.5	52.3	332.6	0.0	0.7	0.0	82.8	18.6	3.0	344.8	112.0
Heilongjiang	101.8	0.0	5862.8	0.0	0.0	3948.9	0.0	164.1	1.6	3735.5	0.3	8.5	18.7	0.0	9.4	0.0	205.3	27.2	14.6	696.1	111.4
Shanghai	0.0	21.0	3.0	0.0	0.0	104.1	0.0	0.0	0.4	1.1	2.2	0.0	1.0	0.4	0.0	0.0	92.9	0.0	0.0	58.8	5.5
Jiangsu	0.0	2412.8	0.0	543.2	0.0	2237.7	0.0	0.0	25.8	194.4	175.3	0.2	88.2	21.0	0.0	0.8	1407.6	209.1	0.0	449.4	183.9
Zhejiang	0.0	103.7	51.9	0.0	86.4	439.6	94.7	45.0	39.1	80.4	96.2	0.0	17.1	4.5	0.0	5.7	644.1	325.7	0.0	272.7	50.8
Anhui	0.0	2822.8	0.0	1160.1	207.4	2190.1	207.7	2.4	62.9	620.5	354.1	1.4	138.9	88.1	1.0	1.9	628.2	138.0	8.2	231.0	152.7
Fujian	0.0	0.2	26.8	0.0	118.6	256.7	253.3	45.6	91.4	29.0	4.9	0.2	67.1	0.0	0.0	4.9	533.4	310.4	52.6	64.6	55.2
Jiangxi	0.0	14.5	35.7	0.0	1279.2	858.6	1366.8	37.0	64.9	102.1	509.1	0.0	162.5	69.0	0.0	14.6	619.3	396.9	25.1	480.1	56.8

Shandong	0.0	4083.9	0.0	4000.1	0.0	108.9	0.0	0.0	102.6	119.5	8.0	1.9	709.2	174.7	0.0	0.0	1462.0	579.4	21.4	315.6	211.6
Henan	0.0	5714.6	0.0	3998.9	0.0	615.0	0.0	0.0	112.7	345.2	155.7	5.8	1151.9	40.0	0.0	2.3	1736.1	442.7	102.8	751.5	326.8
Hubei	0.0	1153.2	794.8	0.0	174.0	1991.7	202.4	203.8	78.9	212.3	971.2	4.3	230.5	204.8	0.0	6.6	1188.6	345.8	36.6	502.4	163.2
Hunan	0.0	28.3	365.8	0.0	1448.2	1291.3	1499.2	75.2	112.2	99.7	1188.9	1.3	106.1	95.7	0.0	7.2	1271.1	500.7	93.1	638.7	124.3
Guangdong	0.0	0.5	121.0	0.0	853.5	0.0	951.9	51.1	148.9	31.2	8.7	0.0	319.1	0.0	0.0	169.2	1227.2	960.5	15.6	329.6	130.0
Guangxi	0.0	3.1	591.2	0.0	810.7	141.0	849.9	55.3	211.9	94.3	20.8	1.0	206.0	1.3	0.0	876.1	1399.7	1174.4	9.9	697.7	108.8
Hainan	0.0	0.0	0.0	0.0	124.5	0.0	122.2	0.1	30.2	2.1	0.0	0.0	32.1	0.0	0.0	21.2	252.9	165.3	0.1	124.0	23.4
Chongqing	0.0	30.1	447.3	0.0	0.0	658.9	0.0	335.0	338.7	96.7	244.3	2.4	62.1	0.0	0.0	2.1	727.2	290.3	30.7	364.1	55.8
Sichuan	10.8	641.9	1863.9	0.0	0.0	1874.9	0.0	684.1	581.8	369.3	1206.2	2.6	261.1	4.4	0.3	9.1	1324.3	693.7	81.3	659.0	137.1
Guizhou	0.0	156.0	1006.4	0.0	0.0	700.5	0.0	699.8	116.0	194.7	516.1	16.1	51.5	1.4	0.4	8.3	1253.1	406.3	143.2	795.9	56.0
Yunnan	0.0	343.7	1763.8	0.0	39.1	802.2	29.3	471.0	56.6	173.1	232.1	4.6	37.5	0.0	0.0	239.9	1084.8	570.0	412.5	1100.6	132.3
Tibet	13.6	25.8	4.9	0.0	0.0	0.9	0.0	1.0	0.0	0.0	19.5	0.0	0.1	0.0	0.0	0.0	23.3	0.0	0.0	165.0	2.6
Shaanxi	0.0	963.1	0.0	1196.9	0.0	105.6	0.0	311.0	35.6	151.9	179.6	29.3	39.5	8.5	0.0	1.4	480.6	1086.9	17.1	543.8	123.1
Gansu	197.4	569.1	1041.0	0.0	0.0	4.0	0.0	565.3	0.0	64.4	198.5	62.5	0.6	19.4	4.3	0.0	337.0	303.3	1.4	687.1	43.2
Qinghai	112.4	0.0	18.9	0.0	0.0	0.0	0.0	86.8	0.0	0.0	153.6	0.0	0.0	0.0	0.0	0.0	43.1	6.2	0.0	140.5	4.7
Ningxia	65.9	57.2	306.3	0.0	0.0	81.1	0.0	118.7	0.0	8.0	1.9	8.8	0.1	0.0	0.0	0.0	118.6	79.6	0.2	365.8	22.8

Xinjiang	408.0	718.8	1019.9	0.0	0.0	74.2	0.0	19.3	4.3	29.5	51.3	152.4	3.6	2217.5	61.1	0.0	302.2	695.4	0.0	824.9	128.2

Table S6. The monthly surface NH_3 concentration of different sub-regions derived from IASI and modeled by our inventory and MEIC inventory (μg N m^{-3}). The R is the correlation of IASI- derived NH_3 vs. simulated NH_3 by our inventory and IASI-NH_3 vs. simulated NH_3 by MEIC inventory.

	HHH			MLYR			NW			NE			SW			SC		
	IASI	This study	MEIC	IASI	This study	MEIC	IASI	This study	MEIC	IASI	This study	MEIC	IASI	This study	MEIC	IASI	This study	MEIC
Jan	5.5	6.3	6.5	3.1	2	2.5	1.8	0.8	0.5	3.9	2.3	2.3	1.1	1.1	1	2.4	1.4	1.1
Feb	10.1	8.2	7.1	4.8	2.8	2.9	2	0.8	0.5	2.9	2	1.7	1	1.2	1.1	2.2	1.5	1.2
Mar	10.1	9.5	5.6	4.5	3.2	2.3	2	0.9	0.4	2.2	2.9	1.4	1.1	1.2	0.9	2.4	1.7	1.2
Apr	12.2	9.7	7.2	6.3	4.8	4.4	2.1	1.3	0.6	2.9	4.2	2	1.2	1.9	1.6	2.8	3	2.4
May	20.8	11.3	7.9	8.3	5.9	5.1	1.8	1.6	0.7	5.5	4.7	2.1	2	2.4	2	3.2	3.8	3.1
Jun	27	12.1	8.6	9.7	6.2	5.5	2.8	1.5	0.9	6.4	4	2.2	1.8	1.9	1.8	3.1	3.8	3.5
Jul	32.1	13.1	8.1	8.1	8.4	5.8	3.6	1.6	0.9	7.9	4.3	2.5	2.4	2.2	1.9	3.3	4.5	3.6
Aug	19.6	10.2	8.3	8.5	7.5	6.8	2.4	1.3	0.9	3.6	3	2.5	1.6	2	2	3.1	4.8	4.2
Sep	15.9	11.8	7.3	3.9	5.5	4.2	1.8	1.2	0.7	2.9	2.5	2.1	1.2	1.7	1.5	2.7	4	3.2
Oct	7.2	12.9	6	3.3	5.3	3	1.5	1	0.5	2.2	2.4	2	0.9	1.5	1.1	1.8	2.8	2.1
Nov	7.4	12	10.4	3.9	5.2	5.7	1.5	0.9	0.7	2.3	1.8	2.3	1	1.4	1.4	2.2	1.9	2
Dec	5.7	8.4	9.8	3.5	3.7	5.3	1.9	0.9	0.7	3.6	2.1	2.8	1.1	1.4	1.4	1.9	1.7	1.8
R		0.57	0.15		0.71	0.63		0.64	0.65		0.7	0.41		0.84	0.81		0.79	0.77

Table S7. The simulation performance of our inventory and MEIC inventory regarding surface NH_3 and $PM_{2.5}$ levels.

Season	This study				MEIC			
	NH_3		$PM_{2.5}$		NH_3		$PM_{2.5}$	
	R	RMSE	R	RMSE	R	RMSE	R	RMSE
Spring (March, April, May)	0.72	3.6	0.5	27.9	0.72	4.1	0.49	26.8
Summer (June, July, August)	0.76	6.2	0.68	36.1	0.67	7	0.65	34.95
Autumn (September, October, November)	0.65	3.1	0.62	31.4	0.61	2.9	0.61	29.8
Winter (December, January, February)	0.6	2.5	0.6	30.4	0.54	2.8	0.59	31.6

Table S8. Livestock number for different provinces of China in 2017 (10000 head)

	Cattle	Other big animals	Goat	Sheep	Pork	Poultry
Beijing	12.8	0.5	8.4	26.8	242.1	3115.2
Tianjin	25.8	0.9	5.3	38.1	297.2	6137.6
Hebei	359.5	28.4	401.4	826.7	3785.3	60637.8
Shanxi	100.7	16.1	382.1	561.1	822.8	12330.3
Inner Mongolia	656.2	168.2	1694.2	4417.8	919.0	10271.1
Liaoning	227.8	63.0	418.8	373.8	2627.2	75891.8
Jilin	337.6	4.5	52.0	347.9	1691.7	42818.4
Heilongjiang	489.3	20.0	175.9	659.3	2090.5	25464.7
Shanghai	6.5	0.0	18.5	1.1	189.7	1272.7
Jiangsu	30.5	2.4	389.3	9.3	2805.5	66150.9
Zhejiang	14.9	0.0	48.4	85.4	1022.4	17319.9
Anhui	80.6	0.3	504.3	0.8	2828.9	87351.9
Fujian	32.6	0.0	89.0	0.0	1606.1	91460.6
Jiangxi	241.4	0.0	95.3	0.0	3180.5	43888.2
Shandong	401.5	12.3	924.4	829.7	5180.7	220172.5
Henan	372.7	3.4	1412.9	269.1	6220.0	90681.6
Hubei	238.0	0.5	543.5	0.0	4448.0	51946.2

Hunan	379.4	1.6	661.7	0.0	6116.3	42263.8
Guangdong	120.7	0.0	93.3	0.0	3712.0	108703.3
Guangxi	326.6	25.8	222.4	0.0	3355.1	86486.3
Hainan	52.8	0.0	68.3	0.1	547.8	15564.9
Chongqing	108.5	2.4	326.7	0.2	1751.1	21315.9
Sichuan	853.2	94.0	1425.9	173.3	6579.1	65259.8
Guizhou	492.4	11.6	362.5	21.0	1825.2	11045.0
Yunnan	810.9	44.5	1149.4	90.8	3795.1	24017.8
Tibet	592.6	35.7	387.6	717.6	19.1	203.4
Shaanxi	151.2	3.3	712.0	156.5	1141.0	5377.6
Gansu	424.3	66.9	398.3	1441.6	682.7	3704.3
Qinghai	546.6	12.7	180.0	1207.4	110.6	520.4
Ningxia	118.3	4.8	99.8	406.8	113.7	1796.0
Xinjiang	433.0	101.3	572.1	3745.8	495.8	9020.7

Table S9. Available mitigation options for croplands in China (derived from meta-analysis of Zhang et al. (2020) and Liu et al. (2021)).

Aspects	Options	Mitigation efficiency
Nitrogen application rate	25% optimal N application rate	18%–32.4%
	50% optimal N application rate	25%–48.5%
	75% optimal N application rate	48.2%–68.3%
Application method	Deep placement of fertilizer	45.1%–79.4%
	Irrigation-fertilization integration management	60.2%–77.4%
Cropland management	Recycling straw to croplands	0%–18.6%
	Reducing basal N fertilizer	26%–62%
N fertilizer type	Urea substitution	8.6%–48.8%
	Application of organic fertilizer	44.7%–63.6%
Enhanced-efficiency N fertilizer	Controlled release fertilizer	46.8%–58.3%
	Inhibitor	21.7%–70.4%

Table S10. Available mitigation options for livestock in China (derived from meta-analysis of Zhang et al. (2020) and Liu et al. (2021)).

Stages	Options	Mitigation efficiency
Feeding	Low crude protein feeding	10%–46%
	Dietary additives	33%–45%
	Phase feeding	~10%
Housing	Floor adaption	10%–50%
	Bedding materials	20%–50%
	Air scrubbing techniques or bio-filter	70%–95%
	Frequent manure removal	25%–30%
	Rapid manure drying	70%–90%
Storage	Solid-liquid separation	20%–30%
	Improvement in storage facility	26%–62%
	Manure surface covers	40%–60%
	Acidification by additives	18%–70%
	Composting	~55%
	Cooling	20%–30%
Spreading	Band spreading	38%–75%
	Incorporation	45%–65%
	Injection (slurry only)	80%–90%
Grazing	Adjusting the grazing time	~10%

Table S11. Agricultural sub-regions of China

Sub-regions	Abbreviation	Province
Huang-Huai-Hai region	HHH	Beijing, Tianjin, Hebei, Henan, Shandong
Middle and Lower Yangtze River region	MLYR	Shanghai, Jiangsu, Zhejiang, Anhui, Jiangxi, Hubei, Hunan
Northwest region	NW	Shanxi, Inner Mongolia, Shaanxi, Gansu, Qinghai, Ningxia, Xinjiang
Northeast region	NE	Liaoning, Jilin, Heilongjiang
Southwest region	SW	Sichuan, Chongqing, Guizhou, Yunnan, Tibet
Southern China region	SC	Fujian, Guangdong, Guangxi, Hainan

Table S12. Total agricultural NH_3 emissions and reduction potential of provinces (Gg)

Province	Agricultural NH_3	NUE-C	OUR	NUE-L	COMB
Beijing	21.89	7.75	2.73	0.99	9.26
Tianjin	38.74	7.65	7.41	2.62	14.74
Hebei	617.37	117.81	128.83	38.76	237.72
Shanxi	201.58	45.42	30.17	19.83	82.21
Inner Mongolia	621.88	79.45	62.14	70.96	183.71
Liaoning	339.19	42.66	44.36	36.60	108.40
Jilin	338.24	14.03	75.28	29.35	112.77
Heilongjiang	400.03	17.35	55.56	35.42	104.67
Shanghai	19.34	6.80	3.86	0.90	9.11
Jiangsu	520.00	165.94	162.56	27.48	285.31
Zhejiang	123.54	50.04	30.49	8.65	69.36
Anhui	460.64	87.88	111.15	29.83	194.22
Fujian	200.35	60.07	33.55	29.80	98.94
Jiangxi	267.73	21.02	32.00	34.09	79.31
Shandong	837.44	71.59	136.83	89.47	270.97
Henan	973.60	142.53	259.58	79.29	417.51
Hubei	507.64	122.35	116.37	45.79	232.86
Hunan	513.96	82.68	72.59	63.61	188.16
Guangdong	422.05	131.94	85.02	60.04	221.37
Guangxi	418.00	60.18	61.90	70.09	168.85
Hainan	76.88	24.35	14.25	11.95	39.94
Chongqing	188.82	46.43	25.05	23.23	78.96
Sichuan	694.36	77.33	62.53	107.32	221.49
Guizhou	268.66	50.91	8.17	43.86	96.89
Yunnan	553.91	119.55	62.25	81.63	221.76
Tibet	165.81	2.28	0.32	38.39	40.69
Shaanxi	335.44	162.91	87.55	20.55	208.70
Gansu	271.62	46.60	3.15	43.43	92.03
Qinghai	149.81	3.92	0.00	33.68	37.69
Ningxia	98.96	34.66	12.48	7.53	45.21
Xinjiang	530.04	145.57	71.00	59.96	229.46

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